PATENT
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

DAVID S. BROWN and
RICHARD E. ROBERTSON

Serial No. 09/832,070

Filed April 10, 2001

NICKEL-CONTAINING ETHYLENE OLIGOMERIZATION CATALYSTAND USE THEREOF

Group Art Unit: 1755

Examiner: James W. Pasterczyk

August 6, 2004

COMMISSIONER FOR PATENTS
P. O. Box 1450
Alexandria, VA 22313-1450

Sir:

SECOND DECLARATION UNDER RULE 132

David S. Brown, the undersigned, hereby declares:

THAT he is one of the coinventors of the above-identified United States patent application; and

THAT he has reviewed the literature article by Xue and believes that that paper and the present application teach fundamentally different points; that Xue teaches that the addition of a second phosphine to the oligomerization system leads to a decrease in K-factor (lighter product distribution), while the present application teaches that the addition of a phosphite with a concomitant reduction in a concentration of the active ligand leads to a system that gives the same activity and K-factor; and

THAT in 1989 he received a Bachelor of Science degree in Chemistry, Mathematics and English from Wake Forest University, and in 1993 he received a Ph.D. degree in Chemistry from the University of California, Berkeley; and

THAT since 1997, he has been employed by Shell Chemical LP and has worked on the Shell Higher Olefins Process and has supervised laboratory work related to the oligomerization of ethylene using nickel catalysts; and

THAT there are differences in the ligand:phosphine or phosphite molar ratio between the invention claimed in the amended claims of the present application and the ratio used in the literature article by Xue; and that the amended claims recite a ligand:phosphite molar ratio of about 50:1 to about 1000:1 (272:1 in the example), whereas Xue uses a ratio of 1:1; and

THAT the use of a higher ligand:phosphite or phosphine molar ratio makes a great difference in the distribution of olefin products that are obtained and in the purity of the desired product; and

THAT the attached table compares the results achieved by Xue in the article, the results from a commercial process, and the results of two experiments that were performed under my direction in order to compare the results at lower ligand:phosphite ratios like those used by Xue with the results at the higher ratio claimed in the amended claims; and that these two experiments were carried out according to the procedure described in Example 1 of this patent application except that the amounts of triethylphosphite and o-dihydrocarbylphosphinobenzoic acid used were changed so that entry 3 was carried out with the 1:1 ligand:phosphite ratio of Xue and entry 4 was carried out with a ligand:phosphite ratio of 50:1 (the low end of the range in the amended claims); and that entry 1 contains the data from example 6 of the Xue literature article and entry 2 contains the data from example 3 of the Xue literature article; and that entry 5 shows representative results achieved in the commercial SHOP (Shell Higher Olefin Process) process, which is different from the present process but produces high quality product; and

THAT the data shows that the use of a small amount of the triethylphosphite compared to the amount of the ligand (entry 4) gives a high product quality (as measured by 1-hexene purity) that is comparable to the results achieved in the commercial process (entry 5) – 98.7 and 96.4 weight percent, respectively; and that the data shows that the use of a larger amount of triethylphosphite compared to the amount of ligand as in the examples from the Xue article (entries 1 and 2) and the comparative triethylphosphite example (entry 3) gives a less desirable product quality of 92.4, 82.57, and 91.5 weight percent, respectively; and

THAT the data shows that the use of a small amount of the triethylphosphite compared to the amount of the ligand (entry 4) gives a more advantageous product distribution (less 1-butene and more total $C_{6-10+}$); and that this is apparent by comparing the results for entry 4 against the results for entries 1 and 2 (from the Xue article) and for entry 3 (which was carried out with the same ligand:phosphite ratio as entries 1 and 2) wherein entry 4 produces only 26.9 weight percent of 1-butene and in entries 1 through 3, 49.45, 86.41, and 71 weight percent, respectively, of 1-butene was produced; and that in entry 4 a total of 73.1 weight percent of $C_{6-10+}$ was produced, whereas in entries 1 through 3, only 50.55, 18.59, and 29 weight percent, respectively, of the $C_{6-10+}$ were produced; and

THAT based on the data shown in the attached table, it is my opinion that the 50:1 to 1000:1 ligand:phosphite ratio of my invention as presently claimed is the reason why my invention achieves better results than are achieved when following the teachings of the Xue article.

David S. Brown further declares that all statements herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date 8-5-04

David S. Brown

| Entry | Experiment | Ligand/Ni Ratio | Phosphite/Ni Ratio | Ligand/Phosphite Ratio | C4 | C6 | C8 | C10+ | 1-Hexene |
|---|---|---|---|---|---|---|---|---|---|
| 1 | Xue, Table 1, Ex. 6 | 1:1 | NA | NA | 49.45 | 28.31 | 10.92 | 11.32 | 92.4 |
| 2 | Xue, Table 2, Ex. 3 | 1:1 | 1:1 | 1:1 | 86.41 | 12.24 | 1.06 | 0.29 | 82.57 |
| 3 | TEP run, high TEP | 1:1 | 1:1 | 1:1 | 71 | 21 | 5.7 | 2.3 | 91.5 |
| 4 | TEP run, low TEP | 1:1 | 0.02:1 | 50:1 | 26.9 | 22.6 | 16.9 | 33.6 | 98.7 |
| 5 | Normal SHOP run | 0.5:1 | NA | NA | 9.3 | 10.6 | 10.7 | 69.4 | 96.4 |